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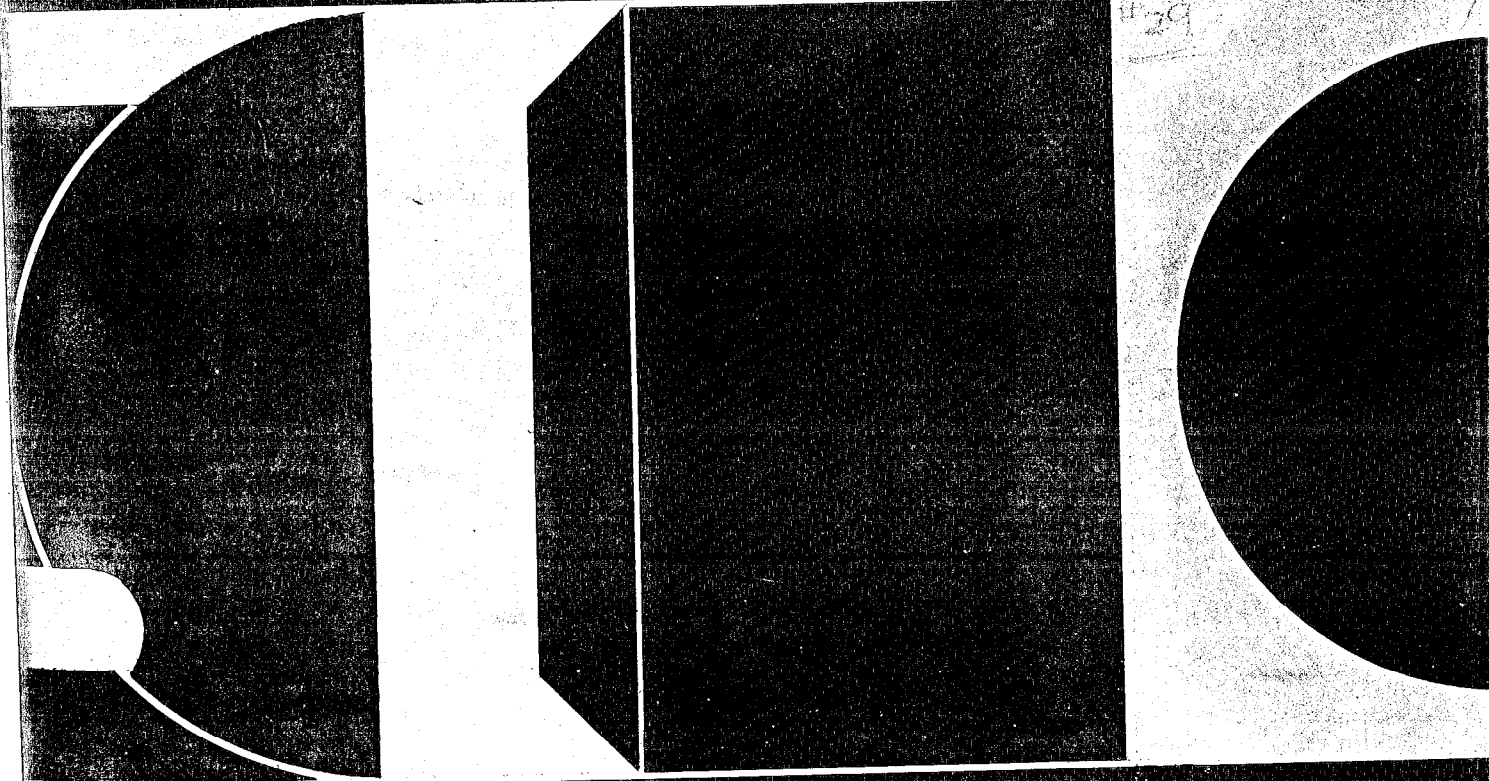
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GAMMA-RAY ASSAY OF LOW-ENRICHED URANIUM WASTE

INSTRUMENTS

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A method based on the detection of 0.767- and 1.001-MeV gamma rays from ^{234m}Pa has been developed for the assay of ^{238}U in large containers of uranium waste. Detailed calibration and assay procedures were obtained for $3\frac{1}{2}\text{-} \times 4\text{-} \times 4\text{-ft}$ plywood boxes of combustible waste. The gamma rays were detected with a large NaI crystal and a line-source "standard" box was used for calibration. The calibration was extended over a wide range of box weights using Monte Carlo calculations of gamma-ray attenuation. The error in the calibration is $<6\%$ (2 σ); much larger assay errors can result from heterogeneities in the waste and from the age dependence of the ^{238}U daughters. The detection limit for a 5-min count is about 30 g ^{238}U in a typical box of combustibles.

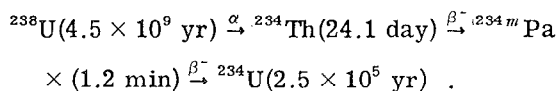
Data generated for the box problem, together with additional Monte Carlo calculations, were used to devise a simple analytical model applicable for the assay of boxes and cylinders in a range of practical geometries. The essential feature of this model is a flux buildup factor which accounts for Compton-scattered photons.

INTRODUCTION

Measurement of the fissionable-material content of waste from the manufacture of nuclear fuels facilitates efficient screening for burial, criticality safety in recovery procedures, and materials accountability. In the case of waste from low-enriched uranium operations, measurements are frequently complicated by the large

size of the containers, typically 55 gal or larger. In particular, the conventional assay based on detection of 186-keV gamma rays from ^{235}U suffers because the gamma-ray attenuation can be severe and difficult to evaluate.

This paper describes a method based on detection of the 0.767- and 1.001-MeV gamma rays from the decay of ^{234m}Pa for the assay of waste. The decay chain which populates ^{234m}Pa is



Quantitative results can be obtained if the activity of this ^{238}U daughter is in equilibrium, which requires an aging period of three months or more, or if the age after the purification of the uranium is known. The equilibrium-emission rates of the 1.001-MeV gamma ray and the cluster of gamma rays near 0.767 MeV are ~ 100 and ~ 60 γ/sec per gram ^{238}U , respectively.^{1,2} Because of this low specific activity, a large NaI detector is needed to obtain sufficient sensitivity for this application.

Although this method does not give a direct assay of ^{235}U , it can be derived from the ^{238}U assay if the enrichment is known or if a nominal value may be assumed. The principal advantage of the method is the penetrability of the high-energy gamma rays, their mean-free-path in combustible waste (with a density of $\sim 0.2 \text{ g/cm}^3$) being about 30 in. Furthermore, since their mass absorption coefficients are essentially constant over the range of medium- and low- Z materials, corrections for self-absorption can be estimated on a weight basis if the waste materials are reasonably homogeneous.

The ^{238}U assay method has been fully developed for one practical example, a $3\frac{1}{2}\text{-} \times 4\text{-} \times 4\text{-ft}$ plywood box, similar to one of the container types used by the General Electric Company, Wilmington, North Carolina, for the temporary storage of

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combustible waste. The calibration and measurement procedures used for this problem are generally applicable to assay problems involving large samples. Gamma-ray attenuation was calculated as a function of box weight with the Monte Carlo gamma-ray transport code MCG.³ Similar calculations were also performed for a 55-gal drum, another widely used waste container. The Monte Carlo results were used to derive flux buildup factors which account for Compton scattering of gamma rays within the container. These data led to a single expression for the buildup factor for boxes and cylinders in a range of practical assay geometries. Finally, a simple analytical model containing the buildup factor was devised for the interpretation of measurements made with this assay method.

DEVELOPMENT FOR LARGE BOXES OF WASTE

Experimental Arrangement

The detection system used for the development of the method for assay of ^{238}U in $3\frac{1}{2}\text{-}\times\text{4-}\times\text{4-ft}$ plywood boxes consists of a 5- \times 5-in. NaI detector enclosed in a 2-in.-thick lead collimator, as shown in Fig. 1. The collimator aperture of this detector is a compromise to achieve both good background shielding and the wide viewing angle needed for large samples. The detector is linked to a commercial portable electronic unit (Eberline SAM II) which is comprised of a power supply, amplifier, scaler, single-channel analyzer, and timer. This particular system, shown in Fig. 2, was originally designed for an experiment to evaluate holdup in a gaseous diffusion enrichment plant.^{4,5}

A "standard" box was designed to simulate uniform distribution of matrix and uranium without the expense and handling problems of a large

box filled with synthetic, uranium-contaminated combustible waste. Photographs of this box, referred to as the line-source (LS) standard box, are shown in Figs. 2 and 3. Three aluminum cylinders, into which 7-in.-diam cans of synthetic waste standards may be inserted, are secured inside a plywood box with cables. Rags (650 lb) packed in the box simulate combustible waste ($\rho = 0.2 \text{ g/cm}^3$). Five standard cans, containing a total of 750 g of uranium (3% ^{235}U) thinly dispersed in shredded Kimwipes (packed to a density of $\sim 0.2 \text{ g/cm}^3$), were used for this study.

Figure 4 shows a plan view of the experimental configuration with the LS standard box. To minimize gamma-ray self-absorption, measurements are made with the detector viewing across the thinnest dimension of the box; i.e., $3\frac{1}{2}$ ft. Effects of inhomogeneities are minimized by (a) locating the detector at a distance from the box which is large compared to its thin dimension, in this case a spacing of 80 in. between the NaI crystal and the near side of the box, and (b) averaging two measurements made with opposite sides of the box facing the detector; i.e., 180-deg rotation of the box. A gamma-ray energy window from 680 to 1120 keV was selected for counting with the single-channel analyzer on the portable electronic unit. This window includes not only the uncollided gamma rays, but also some of those Compton scattered by the matrix. The effect of Compton scattering is evident in Fig. 5, which shows pulse-height distributions of gamma rays from uranium standards in the center of the standard box both with the matrix of rags and without the rags. A significant number of photons scattered out of the full-energy peaks at 767 and 1001 keV by the matrix falls within the counting energy window used for the assay of boxes. Pulse-height distributions of the gamma rays from the uranium standards taken with a Ge(Li) detector showed that

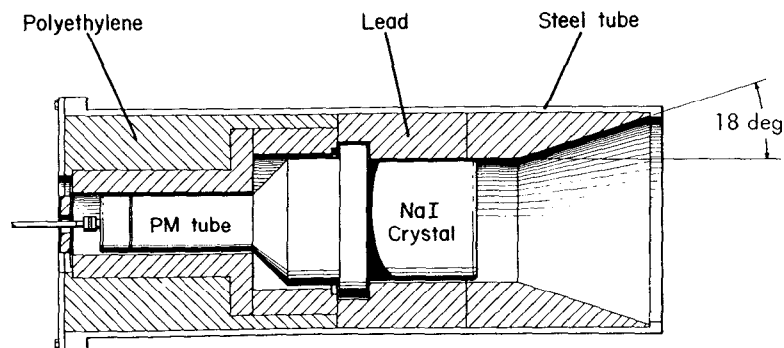


Fig. 1. Details of the 5- \times 5-in. NaI detector and shield assembly. The thickness of the shield nearest the NaI crystal is 2 in.

no contaminating activities were present to interfere with the NaI measurements.

The directional sensitivity of the NaI detector was measured with a ^{137}Cs source, and the results, the collimation function versus angle, are presented in Fig. 6. The angle noted in this figure is that subtended by the half-height of the box at the center of the box. (Referring to Fig. 4, this angle is $\text{arc tan } 24/101$.)

Calibration

Development of the assay method for boxes included the following steps: calibration using the LS standard box loaded with a typical weight of combustible material and measurement of the gamma-ray attenuation of this loading; compar-

ison of experimental results with Monte Carlo gamma-ray transport calculations; and Monte Carlo calculations of the attenuation correction factor versus box weight to provide calibration for a wide range of box weights.

For this development the boxes to be assayed are assumed to be completely full of homogeneous combustibles with gamma-ray attenuation properties of CH_2 ; consequently, the linear gamma-ray coefficients used for the calculations are proportional to the net weight, which is relatively easy to measure under field conditions. If the absorption coefficient of the box contents differs from that of CH_2 , the calibration curves for CH_2 can be adjusted accordingly. Furthermore, the gamma-ray absorption by the uranium is assumed to be zero, since it is negligible for uranium loadings < 5 kg.

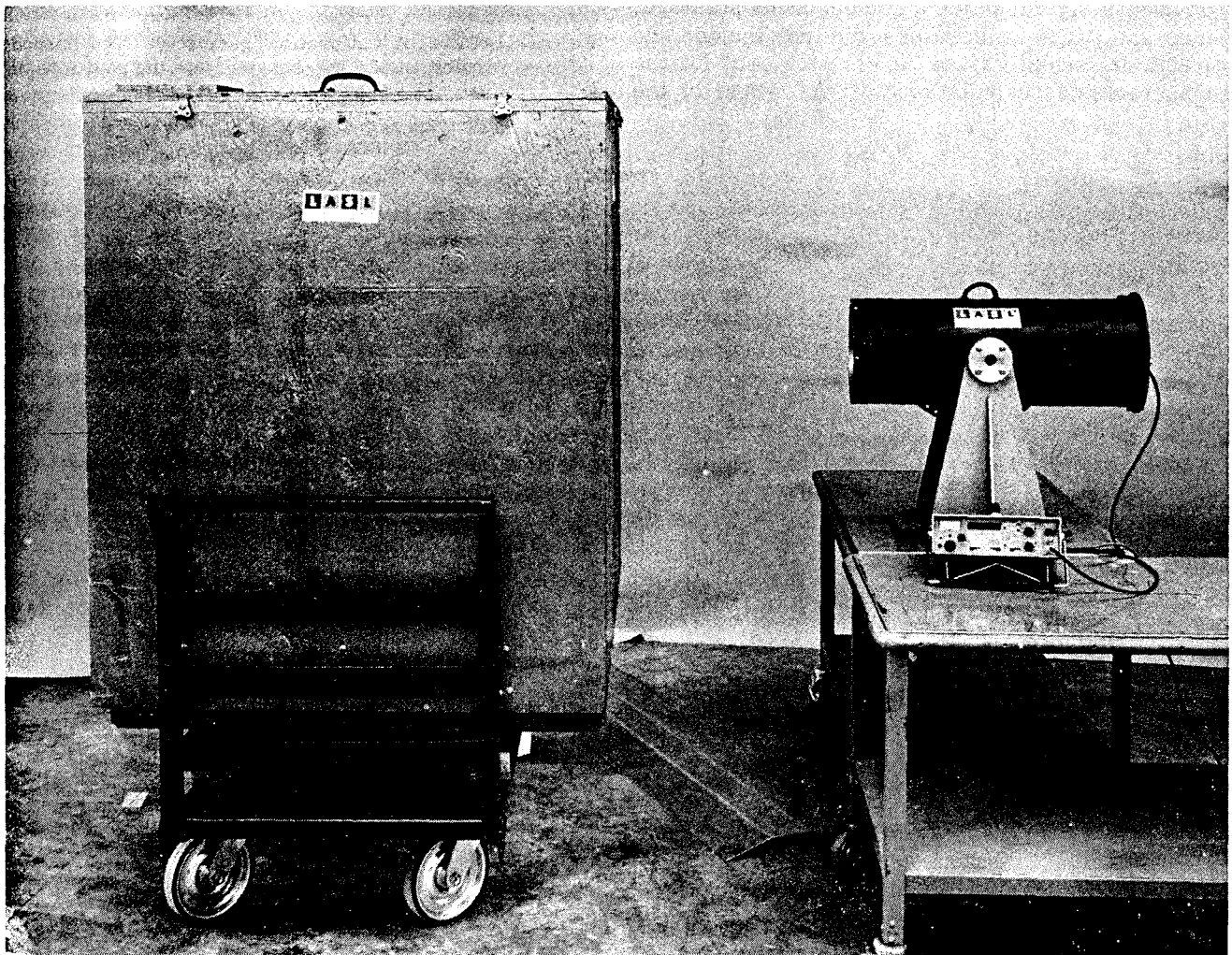


Fig. 2. Detector and electronics (on the right) and the standard box (on the left).

Measurements were performed with the five cans stacked vertically in each of the five positions available with 180-deg rotation of the box, and the calibration was obtained by averaging these results. The gamma-ray attenuation correction factor for the LS standard box, CF_1 , was obtained by taking the ratio of the calibration for the empty box to that for the box filled with rags. Corrections for the self-absorption of the standard cans were applied to the data for the empty box.

The Monte Carlo code was used to calculate gamma-ray fluxes incident on a point isotropic detector located relative to the box, as shown in Fig. 4. From the resulting energy spectra, the uncollided and the Compton-scattered photons could be tallied separately for any energy window. The Monte Carlo results (referred to as MCG) for a homogeneous standard (650-lb) box and for the LS standard box showed the flux from the latter to be higher by only 1%; hence, the LS standard box adequately simulates a homogeneous standard. Since the MCG calculations did not include the directional sensitivity of the detector used for the measurements, calculations of the effect of the

angular response were performed numerically. A detailed discussion of these calculations is presented in the Appendix. This effect reflects an increase of 7% in the flux from the LS standard relative to that from an equivalent homogeneous

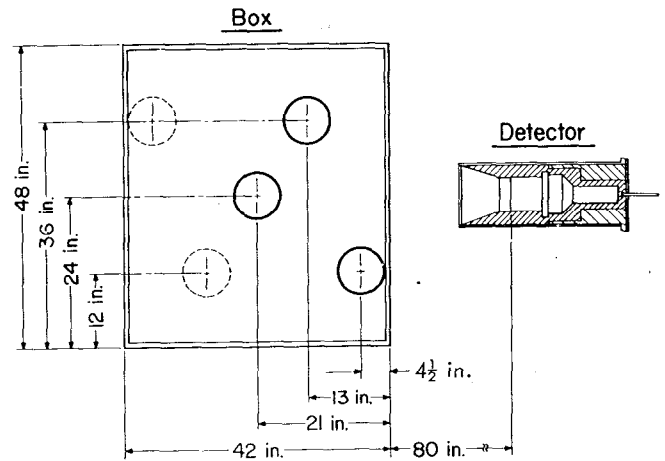


Fig. 4. Details of calibration geometry. The dashed circles inside the box indicate the positions of the holes when the box is rotated 180 deg.

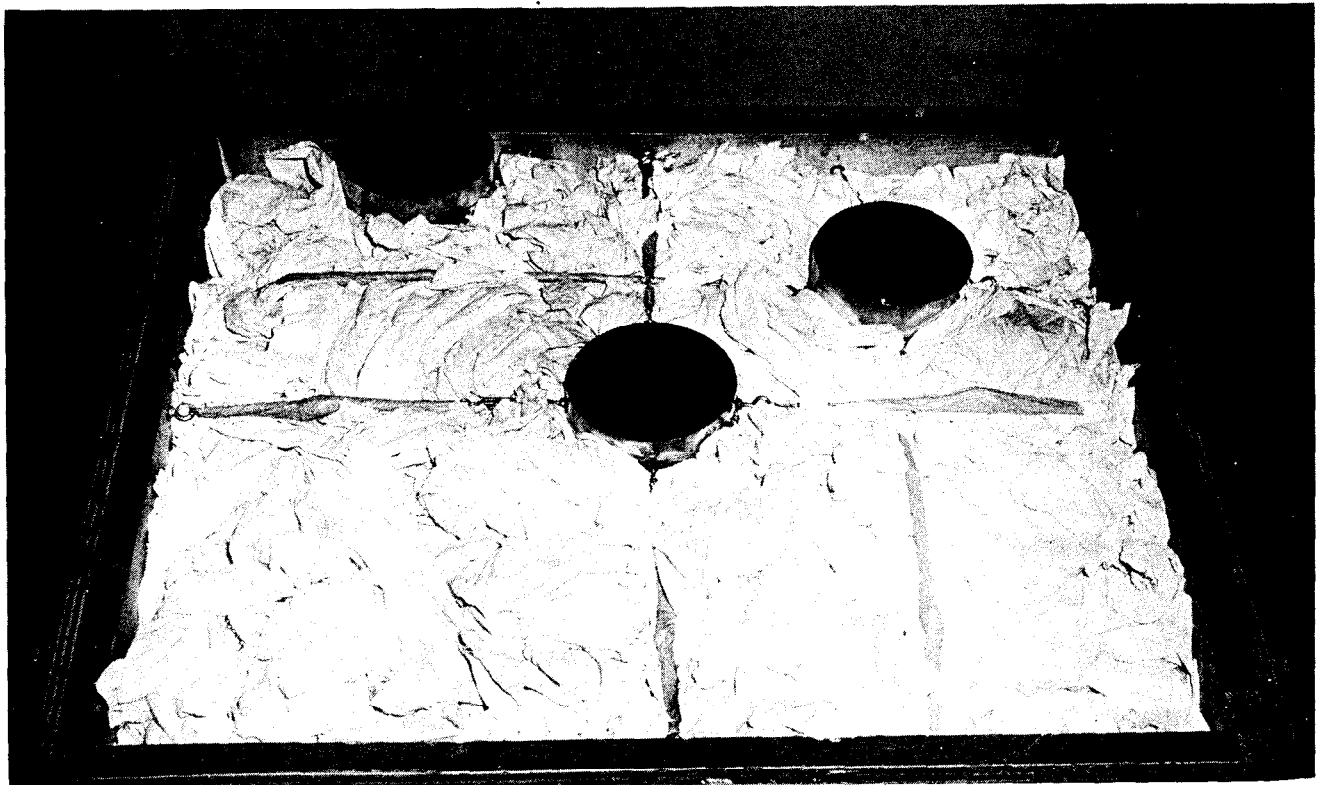


Fig. 3. Top view of the standard box used for calibration for the assay of $3\frac{1}{2}$ - \times 4- \times 4-ft plywood boxes of low-enriched uranium waste. Seven-inch o.d. cans of synthetic uranium standards (not shown) are inserted in the holes. Rags are used to simulate combustible-waste matrix materials.

box. Thus, the total correction required when using the LS standard box instead of a homogeneous standard is 8%.

The complete formula for obtaining the ^{238}U mass in an unknown box, M_x , from the observed counting rate, R_x (count/min), is

$$M_x = 1.08 M_l \frac{R_x}{R_l} \frac{CF_x'}{CF_l'} \quad (1)$$

where

M_l = ^{238}U mass in the LS standard box

R_l = measured counting rate for LS standard box

CF_x' = Monte Carlo result for the attenuation correction factor of the homogeneous unknown obtained from box weight

CF_l' = Monte Carlo result for the attenuation correction factor of a homogeneous box with the same weight as the LS standard.

The measured value of R_l/M_l was 720 count/min per ^{238}U /sec, and the background (at sea level) was ~ 300 count/min.

Figure 7 shows the correction factor measured with the LS standard box, CF_l , versus the lower level of the energy counting window, the upper level being fixed at 1120 keV. For comparison, results obtained from MCG flux calculations for a homogeneous box with 650 lb of CH_2 are presented. Since the gamma-ray attenuation correction factor is a ratio of fluxes, the effect of the angular response of the detection system on this comparison is negligible. The MCG data shown in this figure should be lowered by $\sim 2\%$ to account for the difference in the attenuation coefficients for CH_2 and rags (cellulose), the latter being more like CH than CH_2 in this respect. Further-

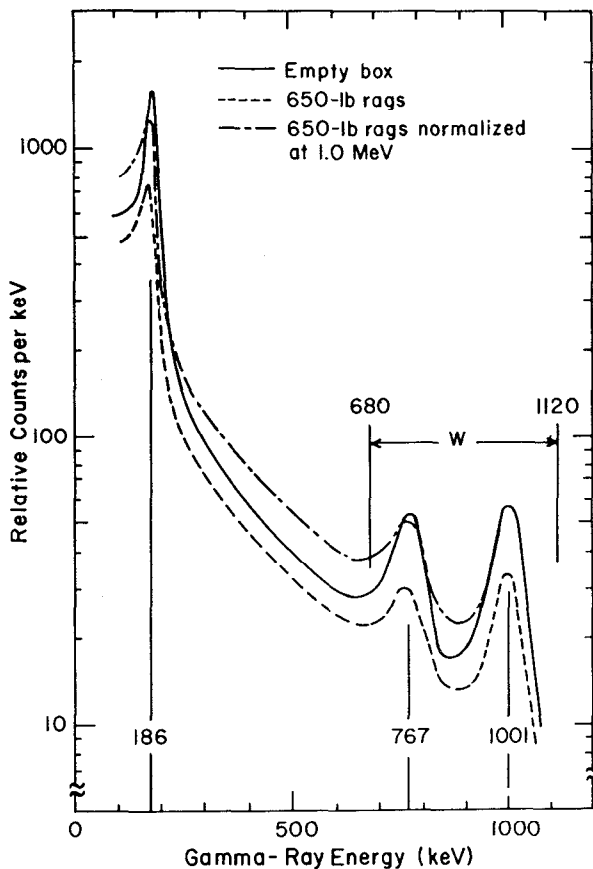


Fig. 5. Pulse-height distributions of gamma rays from uranium standards in the center of the standard box, both with matrix of 650 lb of rags ($\rho = 0.2 \text{ g/cm}^3$) and without rags. To illustrate the effect of Compton scattering by the matrix, the spectrum obtained with the matrix was normalized at 1 MeV to the spectrum without the matrix.

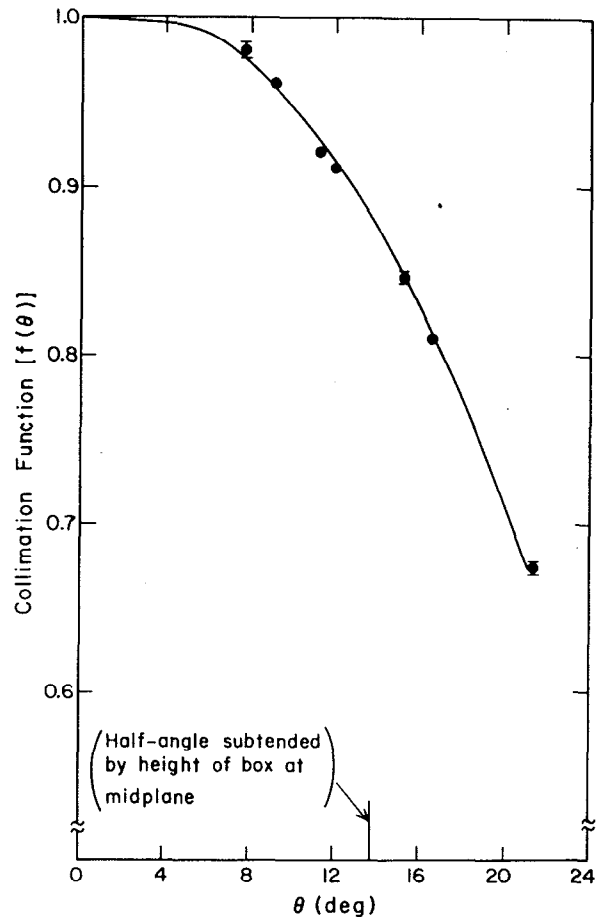


Fig. 6. Directional sensitivity of the detector. Also noted on the plot is the half-angle subtended by the height of the box at the midplane (of the box) which is orthogonal to the detector axis.

more, the effects of the detector gamma-ray response, i.e., the absolute efficiency and the peak-to-Compton ratio, have been neglected for this comparison. These effects should not be significant for energy bias settings ≥ 680 keV because the full-energy detection efficiency does not vary much over this energy range, and the overlap of the Compton response of the 1.001-MeV gamma ray is not large, the Compton edge being at 797 keV. The agreement between the MCG calculations of CF and the measurements is consistent with the statistical uncertainties of the measurements and calculations.

Results of Monte Carlo calculations of the attenuation correction factor of a homogeneous box, i.e., CF' versus box weight (of CH_2), are shown in Fig. 8. Data for two energy counting windows, as well as for the uncollided flux, are presented. The upper abscissa shows values of

$\bar{\mu}L$ corresponding to the net box weights given on the lower abscissa, where $\bar{\mu}$ represents the intensity-weighted average of the linear attenuation coefficients of CH_2 for the 1.001-MeV gamma ray (62%) and the 0.767-MeV cluster of gamma rays (38%), and L is the thickness of the sample in the direction of the detector. The reduction in CF' for the lower biases is caused by Compton scattering in the matrix material. The uncollided flux could be measured directly with a high-resolution Ge(Li) detector.

To show the effect of matrix materials other than CH_2 , calculations were performed for SiO_2 , and the results are compared in Fig. 9. The curves differ because the gamma-ray attenuation coefficient of hydrogen is much larger than those of the other constituents.

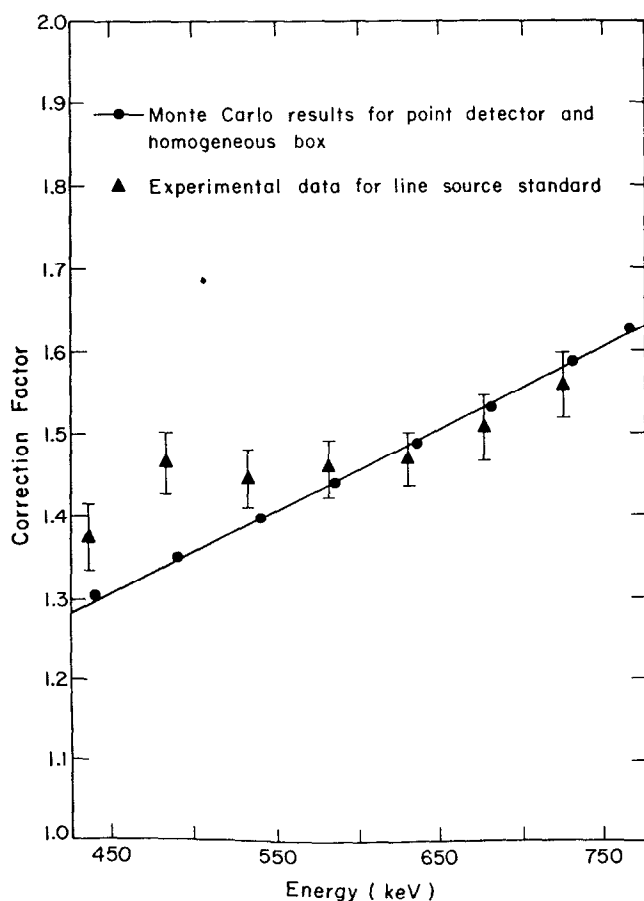


Fig. 7. Attenuation correction factor versus lower level energy window for the standard weight. Results of Monte Carlo calculations are compared with values measured with the standard box. The statistical uncertainties (1σ) of the Monte Carlo results are less than the height of the data symbols.

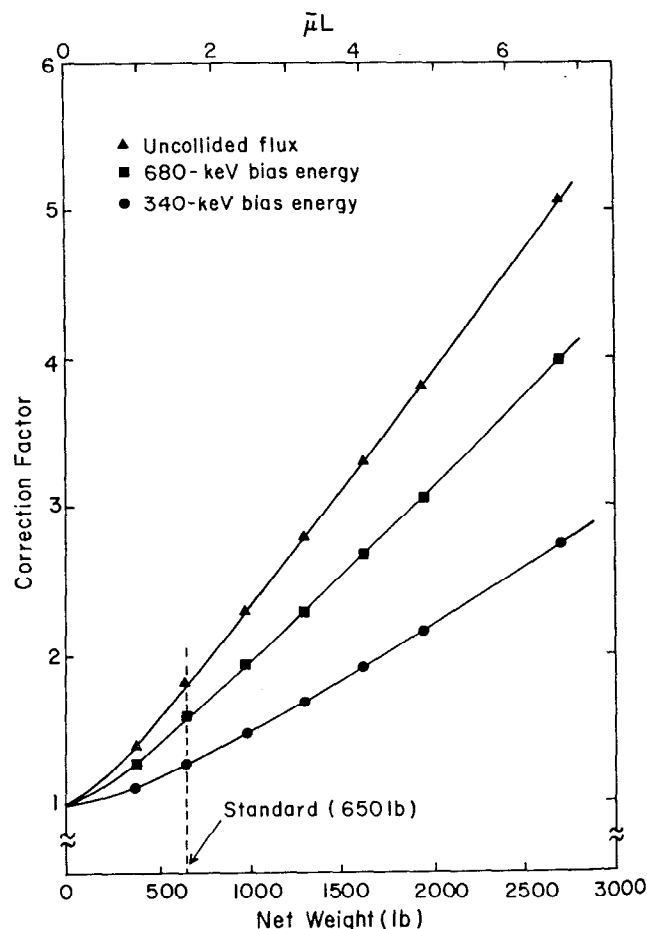


Fig. 8. A plot of Monte Carlo results for attenuation correction factor versus net weight (CH_2) of box for the uncollided flux and for two energy counting windows, both with an upper limit of 1120 keV. The statistical uncertainties (1σ) of the results are less than the height of the data symbols. Note the upper scale for values of $\bar{\mu}L$ corresponding to the box weights.

Sensitivity and Uncertainties

Based on the criterion that a detectable signal is 3σ , where σ is the standard deviation of the background count, the assay system for boxes is capable of measuring quantities of ^{238}U greater than $\sim 30 \text{ g } ^{238}\text{U}$ at sea level in a 5-min count. At Los Alamos, which is 7200 ft above sea level, the minimum detectable mass is $\sim 65 \text{ g}$, reflecting the approximate four-fold increase in cosmic-ray background.

The calibration procedure described above should be accurate to within 6% (2σ). By far the largest errors in this assay method result from derivations from the assumptions used to develop it, namely, homogeneity of the uranium and matrix materials and saturation of the ^{234m}Pa activity. Errors in the measured ^{238}U mass estimated for three examples of heterogeneities are -14% if the uranium is completely surrounded by $\frac{1}{8}$ in. of iron, -10% if all the uranium is located in a few-inch-thick uniform layer at the bottom of the box, and $\sim 55\%$ if the uranium is in the form of individual light-water reactor fuel pellets.

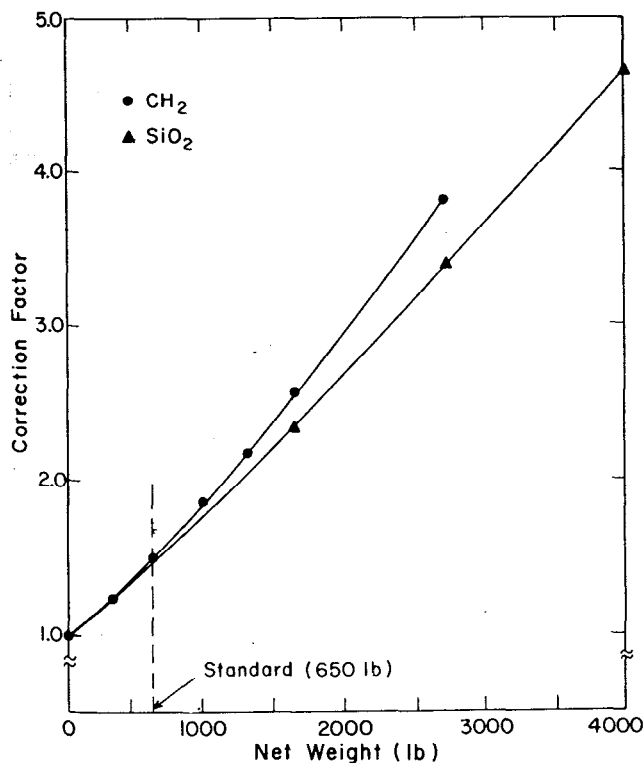


Fig. 9. Attenuation correction factor versus net weight of box obtained from Monte Carlo calculations for CH_2 and SiO_2 . The energy counting window is 680 to 1120 keV. The 1σ uncertainties are less than the height of the data symbols.

A SIMPLE ANALYTICAL MODEL FOR GENERAL APPLICATION

Since Monte Carlo calculations require a sophisticated computer capability and yield results only for specific problems, a simple analytical model, which is more generally applicable but less accurate, has been devised with the aid of the Monte Carlo results.

Calculation of the uncollided flux, Φ_0 , by numerical integration is straightforward; however, obtaining the flux of Compton-scattered photons, Φ_c , by analytical procedure is more difficult. An alternative method for accounting for the Compton flux is to use a flux buildup factor obtained from one or more specific Monte Carlo calculations and defined as

$$B(\bar{\mu}L) = \frac{\Phi_0 + \Phi_c}{\Phi_0} \quad (2)$$

Again, $\bar{\mu}$ denotes the intensity-weighted average of the linear attenuation coefficients of the source gamma rays and L is the sample thickness. Since B is a ratio of fluxes, it should be rather insensitive to exact geometry and, therefore, generally applicable to geometries approximating the far-field geometry, which is defined by a detector-to-sample separation distance, d , much greater than the characteristic container dimension. (The distance for the primary box problem is 80 in., as shown in Fig. 4).

To test the sensitivity of B to geometry, additional MCG calculations were performed for a range of weights of a $3\frac{1}{2} \times 4 \times 4$ -ft box with $d = 40$ in., and a 55-gal drum with $d = 40$ and 80 in. Except for the 55-gal drum with $d = 80$ in., results obtained for $B(\bar{\mu}L)$ for these cases and for the primary box problem ($d = 80$ in.) are shown in Fig. 10. The results for the 55-gal drum with $d = 80$ in. were not discernably different from those obtained for $d = 40$ in. The container wall was included in the thickness, L , and for the 55-gal drum the thickness of the matrix was assumed to be 0.82 times its diameter, D , a recommended scaling for simulation of a cylinder with a slab for $L \lesssim d \lesssim 2L$ (Ref. 6). It can be shown that this scaling factor for the far-field approximation, i.e., $d \gg L$, is $(\pi/4)D$ (Ref. 7).

The maximum spread in the values of the buildup factor for these four cases is about 2.5%, indicating that B is not very sensitive to geometry. Data for the buildup factors were fitted with the function $1 + a[1 - \exp(-b\bar{\mu}L)]$ using the weighted least-squares technique. The resulting parameters are given in Table I. These parameters should not vary with density and Z of the matrix for low- and medium- Z matrices because these are taken into account in $\bar{\mu}$, and for this range of

Z , Compton scattering is the dominant attenuation process. The average values for a and b , 0.308 and 0.531, respectively, are recommended for a single expression for the buildup factor for boxes and cylinders for the NaI counting window used for this study.

Using a narrower counting window reduces the buildup factor but sacrifices counting rate; counting only the photons under the full-energy peaks with a high-resolution Ge(Li) detector essentially eliminates Compton-scattered photons and measures the uncollided flux directly.

Figure 11 depicts a simple one-dimensional model for approximating gamma-ray attenuation correction factors, which is based on the attenuation of the uncollided flux for the far-field homogeneous slab geometry with modifications for $1/r^2$ effects and Compton scattering. In the far-field approximation, the uncollided flux at the detector is proportional to

$$\int_0^L \exp(-\bar{\mu}z) dz,$$

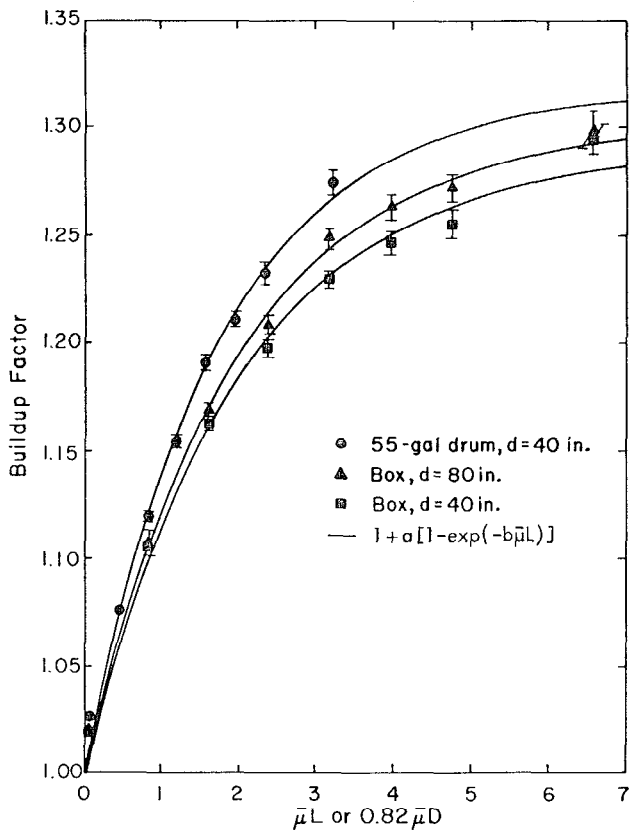


Fig. 10. Flux buildup factor versus $\bar{\mu}L$ obtained from Monte Carlo calculations for three different geometries. For the 55-gal drum the matrix thickness was assumed to be 0.82 times its diameter, a recommended scaling for simulation of a cylinder with a slab. Error bars represent 1σ uncertainties.

giving an attenuation factor of

$$\frac{1 - \exp(-\bar{\mu}L)}{\bar{\mu}L}$$

When d is not much larger than L , $1/r^2$ effects caused by changes in sample attenuation are significant and may be taken into account by assuming $r = d + \delta$, where δ is the position of an equivalent point source in the slab defined by

$$\exp(-\bar{\mu}\delta) \equiv \frac{1 - \exp(-\bar{\mu}L)}{\bar{\mu}L}$$

Thus, in this simple model the effective location of the gamma rays shifts from the center of the slab for $\bar{\mu} = 0$ to the surface facing the detector as $\bar{\mu} \rightarrow \infty$. The flux at the detector with an attenuating medium in the slab, $\Phi(\bar{\mu})$, which includes both the collided and uncollided fluxes, is given by

$$\Phi(\bar{\mu}) = k B(\bar{\mu}L) \frac{1 - \exp(-\bar{\mu}L)}{\bar{\mu}L} \frac{1}{(d + \delta)^2},$$

where k is the proportionality constant. The com-

TABLE I
Fits of Buildup Factors with $1 + a[1 - \exp(-b\bar{\mu}L)]$

	a	b
Box, $d = 40$ in.	0.291	0.493
Box, $d = 80$ in.	0.303	0.508
Barrel, $d = 40$ in.	0.318	0.565
Barrel, $d = 80$ in.	0.320	0.559
Average	0.308	0.531

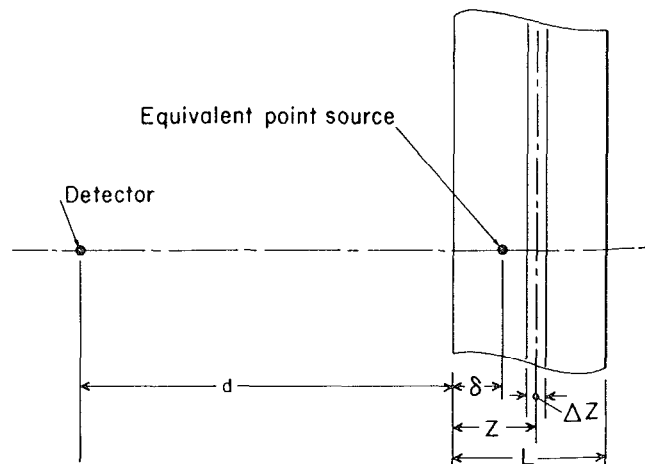


Fig. 11. Geometry of a one-dimensional model for calculating gamma-ray attenuation correction factors. In the far-field approximation, a point source equivalent to the homogeneously distributed source is located at a distance, δ , inside the slab.

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Correction Factor

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Fig. 1

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ponent of flux from Compton scattering is included by means of the buildup factor. Without the attenuating matrix, the flux $\Phi(0)$ is given by

$$\Phi(0) = \frac{k}{(d + L/2)^2}$$

and the gamma-ray attenuation correction factor is

$$CF \equiv \frac{\Phi(0)}{\Phi(\bar{\mu})} = \frac{1}{B(\bar{\mu}L)} \frac{\bar{\mu}L}{1 - \exp(-\bar{\mu}L)} \frac{(d + \delta)^2}{(d + L/2)^2} \quad (3)$$

If $\Phi(0)$ is evaluated by placing the gamma-ray standard inside an empty container, as was done for the box calibration in the present study, the container wall must be taken into account, and the following expression for CF is obtained:

$$CF = \frac{B(\bar{\mu}_w t)}{B(\bar{\mu}_w t + \bar{\mu}L)} \frac{\bar{\mu}L}{1 - \exp(-\bar{\mu}L)} \frac{(d + t + \delta)^2}{(d + t + L/2)^2} \quad (4)$$

where

t, L = wall thickness of the container (i.e.,

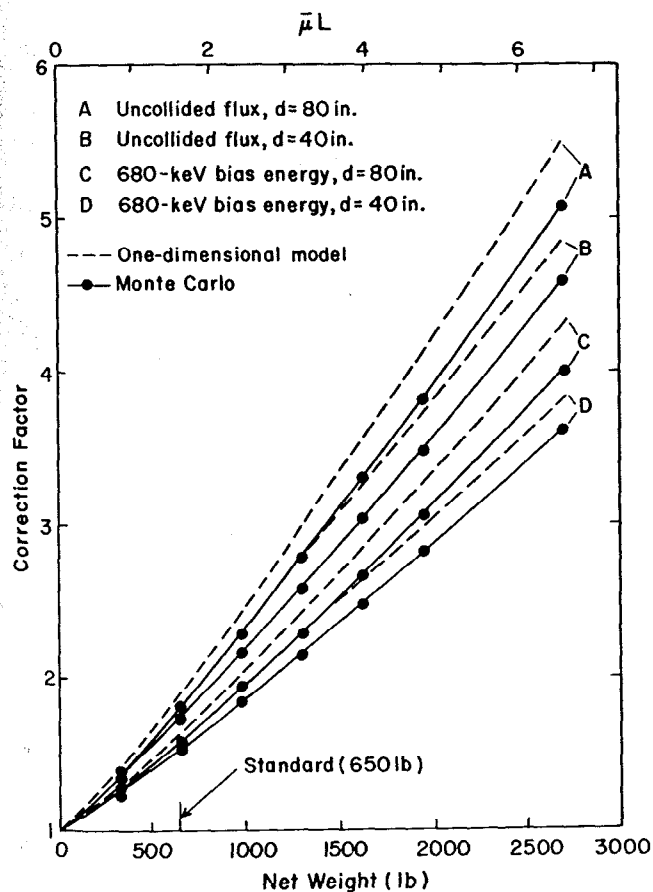


Fig. 12. Attenuation correction factors for $3\frac{1}{2} \times 4$ -ft box with $d = 40$ and 80 in., calculated with a one-dimensional analytical model. Results for both the uncollided and the total flux in the 680- to 1120-keV window are compared with Monte Carlo calculations.

box or barrel) and the thickness of the contents (matrix), respectively

$\bar{\mu}_w, \bar{\mu}$ = linear absorption coefficients of container wall and the matrix, respectively.

Attenuation correction factors obtained with this model are compared in Fig. 12 with MCG calculations. Results for both the uncollided flux and the 680- to 1120-keV window are shown for the $3\frac{1}{2} \times 4 \times 4$ -ft box geometry with distances, d , of 40 and 80 in. The differences between the results of the two calculational methods range from $\sim 3\%$ at 650 lb to $\sim 7\%$ at the heaviest loading and are attributed to the one-dimensional approximation of the model. A similar comparison of correction factors for a 55-gal drum with $d = 40$ in. is shown in Fig. 13. The errors range from -2 to

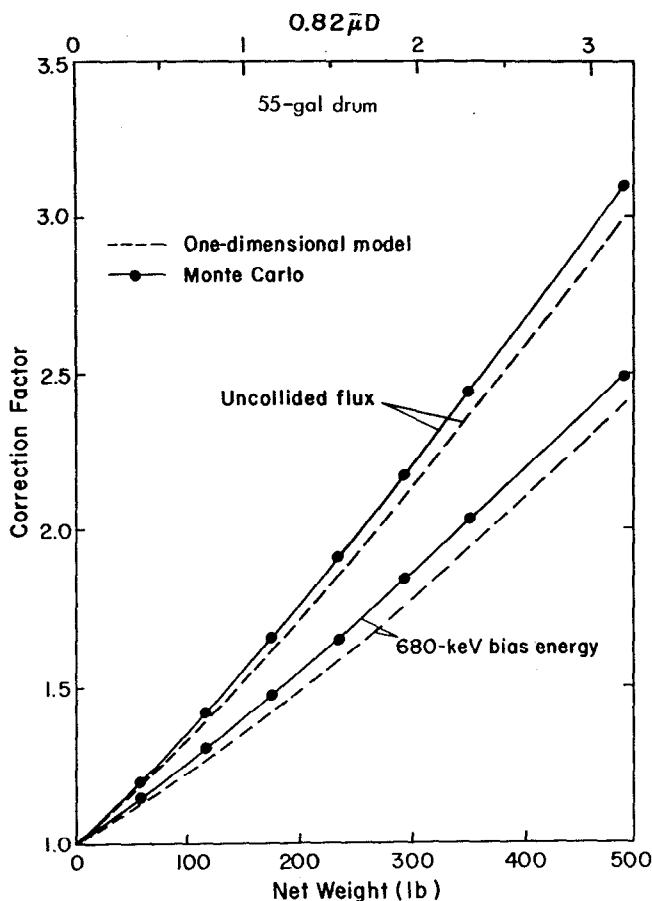


Fig. 13. Attenuation correction factor calculated for a 55-gal drum using a one-dimensional analytical model. Results for both the uncollided and the total flux in the 680- to 1120-keV window are compared with Monte Carlo calculations. The 1σ uncertainties in the Monte Carlo calculations are less than the height of the symbols. Note the upper scale for values of $0.82 \bar{\mu}D$ corresponding to the barrel weights.

-5% over the range shown. Essentially the same agreement was obtained for a 55-gal drum with $d = 80$ in.; these data are not shown in the figure.

CONCLUSIONS

Detection of ^{234m}Pa gamma rays provides a simple, inexpensive means for the assay of uranium waste in large containers. For the range of low- and medium- Z matrix materials, gamma-ray attenuation corrections can be accurately estimated on the basis of the material weight. In practice, assay errors will usually be much larger than the uncertainties in the calibration because of effects of inhomogeneities in the waste and uncertainties in the age after the last chemical separation of the uranium from its daughters.

The procedures described for the calibration and assay of large boxes, in particular the use of an LS standard and the calculation of the effect of the directional sensitivity of the detector, should be generally applicable to a variety of gamma-ray assay problems. The use of Monte Carlo calculations to extrapolate from a single calibration point can reduce the number of physical standards otherwise needed to cover the representative range of loadings. Furthermore, the finding that the flux buildup factor, which accounts for Compton-scattered gamma rays, is almost independent of geometry for most practical choices greatly simplifies the interpretation of measurements with NaI detectors. Once a universal expression for the buildup factor is obtained from Monte Carlo calculations, the uncollided flux can be predicted either by numerical integration or from a suitable analytical model.

In December 1972, the detector and standard box used for this study were shipped to the General Electric Company, Wilmington, North Carolina, for field tests of the method for the assay of plywood boxes of low-enriched uranium waste. General Electric personnel, starting with an earlier, abbreviated version of the calibration and assay procedures presented here, successfully adapted and developed the method for their application and subsequently purchased a similar assay unit.⁸

APPENDIX

CORRECTION FOR DIRECTIONAL DETECTION SENSITIVITY IN THE CALIBRATION FOR HOMOGENEOUS BOXES

To establish the relationship between the experimental calibration with the LS standard box and the MCG results for a homogeneous box and

an isotropic point detector, additional calculations to account for the effect of the directional sensitivity of the detector were required. The measured directional sensitivity, shown in Fig. 6, was used as input data for these calculations.

For this formulation, a monoenergetic source of 1-MeV gamma rays is assumed and multiple scattering processes are neglected; i.e., only the uncollided gamma-ray flux is considered. The following are definitions of the fluxes at the detector position:

$\Phi(\mu)$ = flux from a homogeneous box observed with the directional detector

$\Phi'(\mu)$ = MCG-calculated flux from a homogeneous box with attenuating matrix observed with a point isotropic detector

$\Phi_I(\mu), \Phi'_I(\mu)$ = same as $\Phi(\mu), \Phi'(\mu)$, respectively, except that the box is the LS standard,

where μ is the linear absorption coefficient of the box contents for 1-MeV gamma rays.

The geometry used for the numerical integrations is shown in Fig. A.1. Assuming the gamma-ray source is distributed uniformly within the box, expressions for the fluxes are

$$\Phi(\mu) = k \int_v \frac{f(\theta)\exp(-\mu q)}{4\pi r^2} dV \tag{A.1}$$

$$\Phi'(\mu) = k \int_v \frac{\exp(-\mu q)}{4\pi r^2} dV \tag{A.2}$$

where

dV = volume increment; i.e., $2\pi r dr dz$, as indicated in Fig. A.1

k = gamma-ray source strength per unit volume

$q = z \sec \theta$

r = distance from the volume increment to the detector

$f(\theta)$ = collimation function, or directional sensitivity, as specified by Fig. 6.

If the average values of the collimation function for the empty box, $\overline{f_e(\theta)}$, and for a box with attenuating matrix, $\overline{f_\mu(\theta)}$, are defined as follows:

$$\overline{f_e(\theta)} \equiv \int_v \frac{f(\theta)dV}{\int_v \frac{dV}{r^2}}$$

$$\overline{f_\mu(\theta)} = \int_v \frac{f(\theta)\exp(-\mu q)}{r^2} dV / \int_v \frac{\exp(-\mu q)}{r^2} dV \tag{A.3}$$

then the flux with the directional MCG-calculated

where

The function $\Gamma(\mu)$ is evident from the integration, is the value for $\Gamma(\mu)$ for all boxes

The value for $\Gamma(\mu)$ with the standard for the homogeneous box is

Following the following a

where the sub

Now by ch (A.5) and dividing by the following equation

$$\frac{\Phi(\mu)}{\Phi_I(\mu_s)} = \frac{\Phi'}{\Phi'_I}$$

Fig. A.1. Geometry of the detector box.

then the flux from a homogeneous box observed with the directional detector can be related to the MCG-calculated flux from the homogeneous box by

$$\Phi(\mu) = \Phi'(\mu) \overline{f_e(\theta)} \Gamma(\mu) \quad (A.3)$$

where

$$\Gamma(\mu) \equiv \overline{f_\mu(\theta)} / \overline{f_e(\theta)} \quad (A.4)$$

The function $\Gamma(\mu)$ varies only slowly with μ , or box loading, and has values near unity. This is evident from values obtained for the limits $\mu = 0$ and ∞ . The value for $\overline{f_e(\theta)}$, obtained by numerical integration, is 0.896. For $\mu \rightarrow \infty$, all the gamma rays reaching the detector come from the nearest surface of the box and, by numerical integration, the value for $\overline{f_\infty(\theta)}$ is found to be 0.848. Thus, $\Gamma(\mu)$ for all box loadings has values in the range

$$0.95 \leq \Gamma(\mu) \leq 1$$

The value for $\Gamma(\mu_s)$, i.e., for a homogeneous box with the standard weight, is 0.985. A plot of $\Gamma(\mu)$ for the homogeneous box is shown in Fig. A.2.

Following the same procedures for the LS box, the following are obtained:

$$\Phi_l(\mu) = \Phi'_l(\mu) f_{el}(\theta) \Gamma_l \quad (A.5)$$

$$f_{el}(\theta) = 0.965$$

$$0.97 \approx \Gamma_l(\mu) \approx 1$$

where the subscript, l , is used to denote the LS box.

Now by choosing $\mu = \mu_s$ for the LS box in Eq. (A.5) and dividing Eq. (A.3) by Eq. (A.5), the following equation is obtained:

$$\begin{aligned} \frac{\Phi(\mu)}{\Phi_l(\mu_s)} &= \frac{\Phi'(\mu)}{\Phi'_l(\mu_s)} \frac{\overline{f_e(\theta)}}{\overline{f_{el}(\theta)}} \frac{\Gamma(\mu)}{\Gamma_l(\mu_s)} \\ &= \frac{\Phi'(\mu)}{\Phi'(\mu_s)} \frac{\Phi'_l(\mu_s)}{\Phi'_l(\mu_s)} \frac{\overline{f_e(\theta)}}{\overline{f_{el}(\theta)}} \frac{\Gamma(\mu)}{\Gamma_l(\mu_s)} \end{aligned} \quad (A.6)$$

Since

$$\Gamma(\mu) / \Gamma_l(\mu_s) \approx 1, \quad \overline{f_e(\theta)} / \overline{f_{el}(\theta)} = 0.93,$$

and from MCG calculations,

$$\Phi'(\mu_s) / \Phi'_l(\mu_s) = 0.99,$$

then

$$\frac{\Phi(\mu)}{\Phi_l(\mu_s)} = 0.92 \frac{\Phi'(\mu)}{\Phi'(\mu_s)} \frac{\Phi'_l(0)}{\Phi'_l(0)} = 0.92 \frac{CF'(\mu_s)}{CF'(\mu)} \quad (A.7)$$

In the present context, $\Phi \propto R/M$, where R is the counting rate and M is the ^{238}U mass which is proportional to the gamma-ray source strength. Finally, substituting R/M for Φ gives the calibration equation

$$M_x = 1.08 M_l \frac{R_x}{R_l} \frac{CF'_x}{CF'_s} \quad (A.8)$$

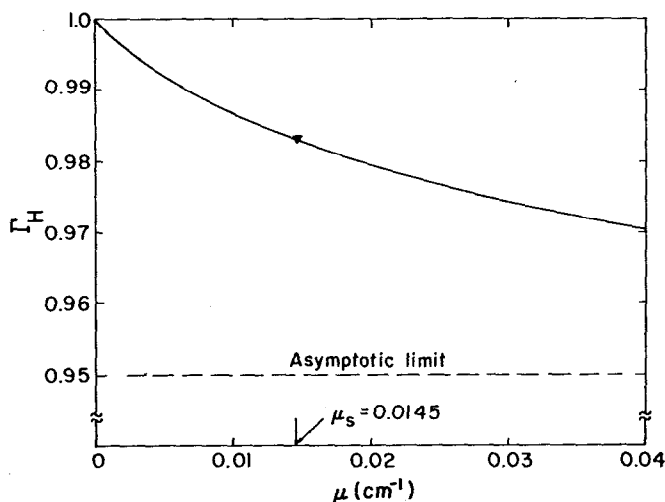


Fig. A.2. The function, Γ , for a homogeneous box versus μ . The point at $\mu_s = 0.0145$ corresponds to the 650-lb loading of the standard box.

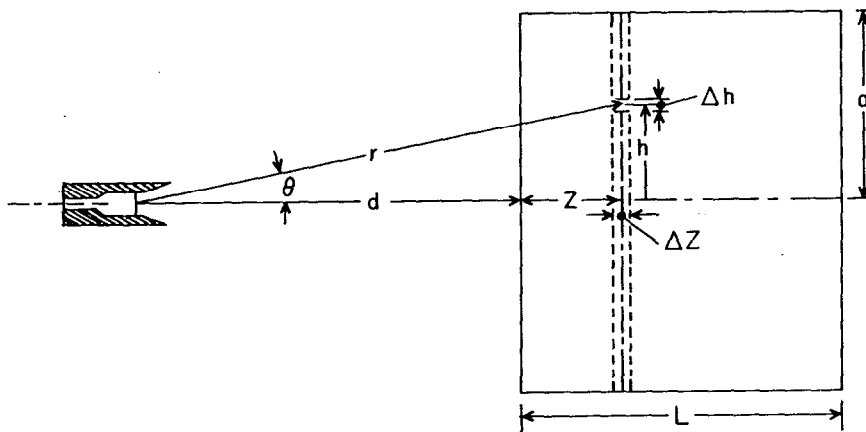


Fig. A.1. Geometry used for numerical integrations to determine the effects of the directional sensitivity of the detector on the calibration for assay of boxes.

where the subscript x denotes the box being assayed and the subscript l refers to the LS standard box. This equation is the same as Eq. (1) in the text.

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REFERENCES

1. A. H. WAPSTRA, "The Decays of ²³⁴Np and ^{234m}Pa(UX₂)," *Nucl. Phys.*, **A97**, 641 (1967).
2. R. GUNNINK and J. F. TINNEY, "Analysis of Fuel Rods by Gamma Ray Spectrometry," UCRL-51086, Lawrence Livermore Laboratory (1971).

3. E. D. CASWELL, J. R. NEERGAARD, C. J. EVERETT, R. G. SCHRANDT, W. M. TAYLOR, and G. D. TURNER, "Monte Carlo Photon Codes: MCG and MCP," LA-5157-MS, Los Alamos Scientific Laboratory (1973).
4. T. D. REILLY, "Assay Experience with MONAL at Oak Ridge," *Proc. 13th Ann. Mtg. INMM*, **I**, 246 (1972).
5. R. B. WALTON, "Nuclear Analysis Research and Development," LA-5091-PR, p. 20, Los Alamos Scientific Laboratory (1972).
6. J. L. PARKER, T. D. REILLY, J. E. FOLEY, R. B. WALTON, and L. V. EAST, "Passive Assay—Innovations and Applications," *Proc. 12th Ann. Mtg. INMM*, **II**, 514 (1971).
7. T. D. REILLY, Los Alamos Scientific Laboratory, Private Communication.
8. C. M. VAUGHAN, L. D. ANSTINE, J. S. DELAP, and J. P. STEWART, "Development, Testing, and Utilization of the Elephant Gun for Measurement of Heterogeneous Radioactive Processing Residuals," *Proc. 14th Ann. Mtg. INMM*, **II**, 292 (1973).

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